

Response to Reviewer #1

We are grateful to reviewer #1 for the valuable and constructive comments that are helpful to improve the manuscript. We have tried to follow all of the suggestions, and make changes accordingly in the main text with red color.

This paper uses OMI SO₂ retrievals to study the effects of emissions and meteorology on SO₂ loading over eastern China during 2005-2016. Monthly OMI SO₂ from BIRA DOAS retrievals are compared with estimated SO₂ emissions from the China Statistical Yearbook. The authors show that OMI-observed SO₂ has decreased significantly over eastern China, particularly for areas with the strongest emissions. They use an EOF analysis to demonstrate that the change is not monotonic and has four phases, with SO₂ increasing during 2005-2007, decreasing sharply during 2007-2008 and 2014-2016, and only slightly increasing or decreasing during 2008-2013. They also show that the emissions and OMI SO₂ are highly correlated over northern part of the country, but less so for southern China. They propose that abnormally dry and stagnant conditions over southern China during 2008-2013 may have caused a slight increase in SO₂ loading, despite continued reduction in emissions. While several studies have examined the recent changes in SO₂ pollution over China using satellite data, this study attempts to provide a somewhat different perspective. The conclusion that meteorology may play a fairly prominent role in the inter-annual changes in SO₂ over southern China is interesting. The paper is well-organized and figures are mostly clear. However, I am not completely convinced that the emission data used can fully support the conclusions drawn in the study. I'd recommend that major changes be made before the paper can be accepted for publication in Atmos. Chem. Phys.

Reply: First of all, we would like to again acknowledge reviewer for the positive comments on our paper. We agree that it is essential to assess the reliability of the emission data used in the study. We have addressed this issue through (i) verification of the sulphur emission statistics released in the official yearbook (OYB), and (ii) repetition of the analyses using another independent inventory.

To verify the OYB inventory and corroborate findings, the Multi-resolution Emission Inventory for China (MEIC) developed by Tsinghua University is adopted. In addition, two other estimates of national annual totals (REASv2 and Zhao) are also used. The references about MEIC, REASv2 and Zhao are listed in the main text.

Overall, the results based on MEIC confirm and reinforce the conclusions obtained by OYB. Firstly, the comparison between the four databases show that, despite the spread in their magnitude, the reported temporal variations are characterized by a similar behavior. We conclude that this justifies the use of OYB as a main source of information. Please see details in Section 2.2 and Figure 1. Secondly, the main conclusions drawn from OYB are not modified when the MEIC dataset is used. This can be judged from the pairwise comparisons listed below and the associated interpretations in the main text.

There certainly exists non-trivial uncertainties related to the currently available emission inventories, especially when considering small spatial or temporal scales or when specific sectors are targeted. Accordingly, we have added a comment in the new Section 6 to highlight current issues and challenges that need to be addressed in the future.

Table pairwise comparison figures between OYB and MEIC

OYB	Figure 2c	Figure 9a	Figure 10	Figure 11
MEIC	Figure 2d	Figure 9b	Figure S3	Figure S4

Specific comments: The authors indicate that the emission data used in this study have very strong seasonal changes in SO₂ emissions from China (almost half of emissions in winter, and only 10% in summer). But this is quite different from many previously published emission inventories which generally suggest a much smaller seasonal change (such as HTAP). Also according to a number of previous studies, the residential sector is in general estimated to contribute roughly 10% of all SO₂ emissions. This is

quite different from what is shown in Figure 9 of this study. The authors may consider using a different, more widely recognized emission inventory for their analysis and check if their conclusion still stands.

Reply: In the sentence “nearly half of the annual totals is released in winter when intensive heating takes place”, the wordings “nearly half” and “released” are not appropriate leading to some confusion. In fact, Figure 3 actually depicts the seasonal variation of SO₂ VCDs rather than SO₂ emissions. In the revised version of the manuscript, we have reformulated the statement as follows: about 35% of the annual totals is taken up by winter, while SO₂ in summer only accounts for 15%; the remaining 50 percent is almost equally divided in between spring and autumn. Please see Lines 228-231.

To explain the pronounced seasonal cycles in SO₂ concentration, an additional figure (Figure 4) has been introduced which correlates the annual cycle of SO₂ VCDs with sulphur emission, precipitable water and temperature at the four hotspots. Although intensive heating during winter in North China raises sulphur release, the variability of the SO₂ emissions alone is not sufficient to drive the pronounced seasonality of SO₂. The remaining variation is associated to seasonal changes in the meteorological conditions. The observed seasonality of atmospheric SO₂ loadings is therefore resulting from variations of both emissions and meteorology. Further details are given in Section 3.2.

In Figure 9 (Figure 11 in the revised version), the Y-axis in a positive direction does not start at zero. We do this because our aim is relative change rather the absolute magnitude. If the origin of Y-Axis is set to zero, the blue bar denoting industrial emission is too tall to recognize the delta change.

It also appears that the emission data used here are on a provincial level (and not gridded) and the authors calculate the emission strength based on the area of each province. Can the authors confirm that? If so, how do the authors calculate total emissions (for example those in Figure 8) for a domain that partially covers several different provinces? Also note that the emissions and SO₂ loading can be quite

inhomogeneous even within the same province.

Reply: We confirm that emission data used here are on a provincial level. Meanwhile, the terms “emission” or “emission amount” always refers to “emission strength”, defined as emitted SO₂ per unit area. To make it clear, we emphasize this in Lines 206-209.

Regional averaged quantities for North or South China are estimated as a weighted average by assigning the district area as a weight. Such explanation is added in Lines 334-336. In fact, the North China and South China are delimited by tracing provinces’ boundaries, so that the North China and South China domains completely cover several different provinces. The demarcation line between the two portions are added in Figure 9.

We understand that emission and SO₂ loading can be quite inhomogeneous within a same region, but this is not considered in this study due to the limitation that only continuous emission data on provinces are gathered at hand. Meanwhile, to match emission data, the gridded SO₂ VCD is aggregated to provincial level. Consequently, the analysis given in Section 4 is constrained to provincial or multi-provincial levels. In Section 6, we particularly point out that future studies should use both gridded SO₂ VCDs and gridded SO₂ emission inventories. Please see Lines 458-462.

It is not clear how the “north” and “south” are defined in this study. One would assume that Cheng-Yu, PRD, and YRD are all part of the “South”. But the SO₂ time series in Figure 6 indicates that they have different trends during 2008-2013. How would the authors explain these different trends when Figure 10 appears to show generally similar meteorological conditions for the three regions?

Reply: Thanks for your suggestion. We added a demarcation line in Figure 9 that separates South China and North China.

We also realize that Figure 6 may not be clear enough to convey the major idea. This is because the monthly anomalies mask the primary signal, and trend is also not a suitable indicator for rebound phenomenon. Therefore we designed a new figure (Figure 8) which presents the temporal evolution of annual SO₂ from 2005 to 2016 in

each province of eastern China, with the segment over 2009-2013 highlighted by red color. It confirms that the SO₂ does not evolve in a monotonic way but shows a striking rebound during 2009 to 2013. This pattern is observed throughout most of the region, with only two exceptions: the Guizhou and Guangdong provinces that experienced a monotonic decrease since 2005. See Figure 8 and detailed explanation in Lines 305-313.

The “sudden downward shift of household emissions” in the south is quite surprising. Do the authors have an explanation for this? Or is this simply indicative of methodology change in the emission inventory?

Reply: We have double checked and can confirm that the OYB inventory shows a sudden downward shift of household emissions for South China. Unfortunately, the official year book doesn't provide any clue whether this shift results from a change in methodology or reflects real changes in emission inventory. As a result, we decided to avoid any speculation in the main text.

We also note that the MEIC inventory does not shown evidences for such an “abrupt-shift” behavior. Nevertheless, both inventories do report a reduction of household emissions in South China, irrespective of the exact manner. The phrase “a sudden downward shift of household emissions” has been removed from the abstract and the conclusions. Instead we refer to “the coordinated cuts of industrial and household emissions”. Please see Lines 39-40 and 438.

Figure 4: there seem to be some negative SO₂ values in the figure? Can the authors confirm that?

Reply: In this study, all negative SO₂ values are eliminated prior to the research. Thus, there are no negative values in Figure 4 (Figure 6 in the revised version). The label bar includes ‘less than 0’ bin, because we expect it to be symmetric. However, no patch on the shaded plot indicates less-than-0 value.

In addition, the YRD and PRD regions are slightly enlarged in the revised manuscript, to mitigate possible uncertainties.

Figure 8: What is the unit for emissions? What does each data point represent in the scatter plot?

Reply: The unit for emission is ton/km², and each data point corresponds to one year and one province. Such key notes are inserted into the figure caption. Please see Figure 10.

Figure 10: which area is the vertical profile in (d) for?

Reply: The area is 23-31°N, 105-122°E rectangle. In addition, we find that the result is not sensitive to the chosen area.

Writing: the authors should also make an attempt to improve the writing. Short, simple sentences in some cases may make the paper easier to follow.

Reply: Thank you very much for your suggestion. Further polishing has been applied in the revised version.

Response to Reviewer #2

We thank reviewer #2 for his suggestions to improve our manuscript. We have done our best to address each of them. The revisions in the manuscript are highlighted by red color.

General comments: In this paper, Ting Wang et al. analyzed the spatial distribution and temporal variation trends of SO₂ VCD and emissions in different regions of China in the last decade based on the OMI observation and emission inventory. Further they discussed effects of meteorological conditions on the SO₂ variations based on the differences of emissions and SO₂ VCDs in South China. In general the scientific topic is meaningful, and the perspective of understanding effects of meteorology on SO₂ depositions and dispersions is novel. However I have two major concerns below:

Reply: We would like to again acknowledge the reviewer for his positive comments on our manuscript.

1) A credible emission inventory is quite a foundation of the study. However the authors do not give a peer-reviewed publication of the emission inventory in Section 2. The authors should cite some papers to introduce the methodology and validation of the inventory. Meanwhile it could be more convincing if the authors do the same analysis based on another available peer-reviewed emission inventory

Reply: Thanks for this valuable suggestion. The official estimate of SO₂ emission inventory is published every year, and it is certain that peer review processes are undertaken before publication although these are not open to the public. The yearbook also briefly introduces the methodology to build the inventory, which has been added in Section 2.2. Unfortunately, no further detailed information is provided in the yearbook.

We understand the crucial concern regarding the reliability of the emission data used. To guarantee the robustness of this study, we have sought to address the concern in the revised version as you suggested. A similar comment was raised by the first referee and

we refer to our corresponding reply.

2) The author did not consider any effects of regional transports in the discussion of the discrepancy of SO₂ VCD and emissions. SO₂ life time could be long and has a big variability. SO₂ could be transported by on an order of 100 km, especially during night.

Reply: Thank you for this comment. The effect of regional transport is of importance, and climate-chemical coupled models are usually applied for its evaluation. Alternatively, we have used an Effective Emission Index (*EEI*) that accounts for the impact from both local and remote sources. Adopting results published by Zhang et al. (2015), we get: for North China, within-region SO₂ emission contribute 68% followed by 19% from South China and 13% from other regions; for South China, within-region emissions provide 66%, while transport from North China and other regions amounts to 17% and 17% respectively. Based on these statistics and assuming that the *EEI* is linearly dependent on *N* and *S* and that external contributions remain fixed, the *EEI* index is formulated. For comparison purpose, we also define an Emission Index (*EI*) that considers single effect from within-in region emission. The detailed procedure to construct the indices are presented in Lines 339-365.

Note that we only consider the inter-regional transport between North China and South China. Because the spatial scale is large, we find that integrating the role of inter-regional transport does not alter the overall pattern and result. However, the best way to unveil fine scale details of transport is using climate-chemical coupled models. Due to the limited resources available for this study, this could unfortunately not be attempted. Future directions are highlighted in Section 6.

Specific Comments:

1) Line 116-117: how significant is the improvement of the new product on the study of variation trends? Do the authors compare the variation trends based on the new product with those based on the previous product?

Reply: Compared to the BRD OMI NASA SO₂ product, the BIRA retrievals proved to be better both in terms of noise level and accuracy. This product also includes a full

characterization (errors, averaging kernels, etc.). The improved OMI PCA SO₂ product of NASA show similar performance and long-term trends as the BIRA product. The BIRA SO₂ product has also been validated in China using long-term MAX-DOAS data (Theys et al., 2015; Wang et al., 2017). As these comparisons have been done before this study, we added a short remark in Lines 125-130.

2) Line 120-121: What kind of background correction is applied? Can the correction cause artifacts of some weak signals of SO₂ in some regions which are dominated by the natural sources as discussed in Line 151-155?

Reply: The correction we use here is based on a parameterization of the background values that are then subtracted from the measurements. The scheme first removes pixels with high SZA ($>70^\circ$) and SCDs larger than 1.5 DU (measurements with presumably real SO₂) and then calculates the offset correction by averaging the SO₂ data on an ozone slant column grid. This is done independently for each across-track position and hemisphere, and the correction makes use of measurements averaged over a time period of two weeks around the measurement of interest. The details of background correction can be found in Theys et al. (2015).

Yes, the low level SO₂ columns are subject to large uncertainties and the background correction is an important source of error. However, the regions with weak SO₂ signals/background SO₂ are not the subject of the present paper.

3) Fig. 3a: The author should explain the line around 40 N latitude with high values in winter

Reply: There is a belt of large positive values extending along 40°N in winter. However, it is a known artefact due retrieval limitations at large solar zenith angles. This does not incur any barrier to subsequent investigations, since our focus is eastern China that does not include this belt. Please see details in Lines 273-275.

4) Line 161 and Fig. 2a: snow could cover the surface in the western and northern part of China in the seasons, except summer. The snow covered surface could impact the

retrievals of SO₂ VCD. This could be the reason of the missing values of satellite SO₂ VCDs in the two regions, especially in winter. Do the authors consider the point in the discussion?

Reply: Following your suggestion, we added Figure S2 and a paragraph to elaborate on this point. Figure S2 is designed to evaluate the availability of monthly SO₂ data relative to the entire period. As mapped in Figure S2, there appears to be a substantial fraction of data gaps in western and northeastern China, especially in the winter half year. This can be attributed to snow cover surfaces and high solar zenith angles, which invalidates the measurability. However, the missing data issues in northeast and western China have virtually no impact on our study, because we mainly focus on the highly polluted eastern China.

Please see Figure S2 and relevant interpretations in Lines 135-145.

Meanwhile in the lines of 161-163, the authors attribute the higher SO₂ amounts in summer than other seasons to the natural emissions. However the snow coverage effect could also play a role.

Reply: In the light of the considerable data gaps in western China as shown in Figure S2, it is impossible to draw firm conclusion. Therefore, we remove this statement in the revised version.

5) Line 159: The authors conclude that “nearly half of the annual totals is released in winter” because of the significant higher SO₂ VCD in winter than in other seasons. However SO₂ lifetime could be also longer in winter. The larger SO₂ VCD values could be also related to longer lifetime of SO₂ due to its easy accumulations in winter.

Reply: Thank you for this suggestion. The lifetime does take an important role in shaping the seasonality of SO₂ VCD. To better explain the pronounced seasonal cycles in SO₂ concentration, an additional figure (Figure 4) has been included to illustrate the annual cycle of SO₂ VCDs and its relation to sulphur emission, precipitable water and temperature at the four hotspots. On the one hand, intensive heating during winter in North China raises sulphur release. However, emission alone is not adequate to explain the pronounced seasonality of SO₂. Temperature and humidity are cold and dry in

winter due to the influence of winter monsoon, which jointly weakens the rate of oxidation and wet deposition. Accordingly, it is expected that SO₂ molecules will have a longer lifetime and will thus accumulate easier, as you suggested. The opposite is true for summer, when chemical reaction is active and wet removal is effective. Please see Figure 4 and relevant interpretations in Lines 233-243.

1 **Spatial and temporal changes of SO₂ regimes over China in**
2 **recent decade and the driving mechanism**

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22 **Abstract:** The spatial and temporal changes of SO₂ regimes over China during 2005 to
23 2016 and their associated driving mechanism are investigated based on a state-of-the-
24 art retrieval dataset. Climatological SO₂ exhibits pronounced seasonal and regional
25 variations, with higher loadings in wintertime and two prominent maxima centered in
26 the North China Plain and the Cheng-Yu District. In the last decade, overall SO₂
27 decreasing trends have been reported nationwide, with spatially varying downward
28 rates according to a general rule—the higher the SO₂ loading, the more significant the
29 decrease. However, such decline is in fact not monotonic, but instead four distinct
30 temporal regimes can be identified by empirical orthogonal function analysis. After an
31 initial rise at the beginning, SO₂ in China undergoes two sharp drops in the periods
32 2007-2008 and 2014-2016, amid which 5-year moderate rebounding is sustained.
33 Despite spatial coherent behaviors, different mechanisms are tied to North China and
34 South China. In North China, the same four regimes are detected in the time series of
35 emission that is expected to drive the regime of atmospheric SO₂, with a percentage of
36 explained variance amounting to 81%. Out of total emission, those from industrial
37 sector dominate SO₂ variation throughout the whole period, while **the role of household**
38 **emission remains uncertain**. In contrast to North China, SO₂ emissions in South China
39 exhibit a continuous descending tendency, due to **the coordinated cuts of industrial and**
40 **household emissions**. As a result, the role of emissions only makes up about 45% of the
41 SO₂ variation, primarily owing to the decoupled pathways of emission and atmospheric
42 content during 2009 to 2013 when the emissions continue to decline but atmospheric

43 content witnesses a rebound. Unfavorable meteorological conditions, including
44 deficient precipitation, weaker wind speed and increased static stability, outweigh the
45 effect of decreasing emissions and thus give rise to the rebound of SO₂ during 2009 to
46 2013.

47 **Key words:** SO₂, China, spatiotemporal regimes, mechanism

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51 **1 Introduction**

52 In recent decade, air pollution has persistently plagued China, especially in leading
53 economic and densely populated areas (Chan and Yao 2008; Ma et al., 2012; Chai et
54 al., 2014). In China, environmental protection agencies identify six major pollutants of
55 concern, including sulfur dioxide (SO₂), nitrogen dioxide (NO₂), ozone (O₃), carbon
56 monoxide (CO), fine particulate matter (PM_{2.5}) and coarse particulate matter (PM₁₀).
57 Then, values of the six pollutants are transformed into a single number called Air
58 Quality Index (AQI) for effective communication of air quality status and
59 corresponding health impact (MEPC, 2012).

60 SO₂ is one of the six major pollutants in China (Ren et al., 2017). It is harmful to
61 human health, affecting lung function, worsening asthma attacks and aggravating
62 existing heart disease (WHO, 2018). It also leads to the acidification of the atmosphere,
63 and the formed sulfate aerosol is one of the most important components of fine particles
64 in cities (Meng et al., 2009). Overall, SO₂ is a key influencing factor for atmospheric
65 pollution, and it poses great threats to life, property and environment (Wang et al., 2014).

66 Compared to airborne and ground-based remote sensing, satellite platforms permit
67 near-global coverage on a continuing and repetitive basis, enabling quick and large-
68 scale estimation of pollution patterns (Yu et al., 2010). Since the world's first weather
69 satellite TIROS-I launched in 1960, satellites have become a crucial part of Earth's
70 observations and practical applications (Yu et al., 2010). Till now, SO₂ has been
71 measured globally by several operational satellite instruments, such as OMPS (Zhang

72 et al., 2017), GOME-2 (Munro et al., 2006; Rix et al., 2010) and OMI (Lee et al., 2011;
73 Li et al., 2013; Theys et al., 2015).

74 With the aid of satellite data, in the past decade, various attempts have been made to
75 explore the variation of SO₂ loadings in China. Lu et al. (2010) report that total SO₂
76 emissions in China have increased by 53% from 2000 to 2006, followed by a growth
77 rate slowdown and the start of a decrease. Li et al. (2010), Yan et al. (2014), and Zhang
78 et al. (2012) all highlight the prominent reduction of SO₂ during 2007 and 2008, as a
79 consequence of the widespread deployment of flue-gas desulfurization and the strict
80 control strategy implemented for preparation of the 2008 Olympic Games. Throughout
81 the past decade, 90% of the locations in China have shown a decline in SO₂ emissions,
82 as highlighted by Koukouli et al. (2016). Such widespread declines are ascribed to
83 effective air quality regulations enforced in China (van der A et al., 2017). Furthermore,
84 Krotkov et al. (2016) and Li et al. (2017) both compared the sulfurous pollution in
85 China and India, and pointed out their opposite trajectories. Since 2007, emissions in
86 China have declined while those in India have increased substantially. Nowadays, India
87 is overtaking China as the world's largest emitter of anthropogenic SO₂. In addition,
88 several studies conducted analyses on SO₂ in sub-regions of China, for example Jin et
89 al. (2016), Lin et al. (2012), Wang et al. (2015) and Su et al. (2011). All these studies
90 contributed to a better understanding of SO₂ changes in China. However, there are still
91 key issues to be addressed. First, with the pace of considerable progress made on SO₂
92 retrieval, updated data products are now available to accurately derive recent SO₂

93 variations in China. Second, although the general decreasing tendency has been
94 revealed, the specific spatial and temporal regimes remain unclear. Does the SO₂
95 decrease monotonically, or is there a complicated oscillation? How similar/different are
96 SO₂ variations in different parts of the country? Third, there is more to be learned about
97 the driving mechanisms that govern SO₂ variations. Previous studies have mainly
98 focused on the impact from amounts of emission. However, the SO₂ content is not only
99 dependent on emissions but also on atmospheric conditions. Therefore, how large is the
100 influence of atmospheric variability on the variation of SO₂?

101 The overall goal of this study is to quantify the spatial and temporal changes of SO₂
102 regimes over China in the last decade and to disclose the driving mechanism, based on
103 a new-generation of SO₂ retrieval dataset (Theys et al., 2015). **Figure S1 labels the**
104 **provinces of China.** The manuscript is organized as follows. Section 2 describes the
105 new SO₂ product, and statistical databases of SO₂ emission and atmospheric data are
106 introduced. In Section 3, we evaluate the general patterns of SO₂ including mean
107 distribution, long-term trends and seasonality. Subsequently, Section 4 identifies the
108 specific regimes of SO₂ variability and the associated driving mechanisms. Finally,
109 concluding remarks are presented in Section 5.

110

111 **2 Data**

112 **2.1 SO₂ VCD retrievals**

113 The Ozone Monitoring Instrument (OMI) is one of four sensors onboard the Aura

114 satellite launched in July 2004 (Levelt, J. et al., 2006). In recent years, Belgian Institute
115 for Space Aeronomy (BIRA) and cooperators have developed an advanced Differential
116 Optical Absorption Spectroscopy (DOAS) scheme to improve the retrieval accuracy of
117 SO₂ in troposphere. A SO₂ vertical column product is generated based on the algorithm
118 applied to OMI-measured radiance spectra (Theys et al. 2015). The retrieval scheme is
119 a based on a DOAS approach, including three steps: (1) a spectral fit in the 312-326 nm
120 range (other fitting windows are used for volcanic scenarios but are not relevant for this
121 study), (2) a background correction for possible bias on retrieved SO₂ slant columns,
122 (3) a conversion into SO₂ vertical columns through radiative transfer air mass factors
123 calculation, accounting for the SO₂ profile shape (from the IMAGES chemistry
124 transport model), geometry, surface reflectance and clouds.

125 Compared to the BRD OMI NASA SO₂ product, the BIRA retrievals proved to be
126 better both in terms of noise level and accuracy. The BIRA product is also fully
127 characterized (errors, averaging kernels, etc.). The improved OMI PCA SO₂ product of
128 NASA show similar performance and long-term trends as the BIRA product. The BIRA
129 SO₂ product has been validated in China with long-term MAX-DOAS data (Theys et
130 al., 2015; Wang et al., 2017).

131 The dataset is made available on a 0.25° and 0.25° regular latitude-longitude grid
132 over the rectangular domain 70-140°E, 10-60°N, and covers the period of 2005 to 2016
133 at monthly interval. In addition, a cloud screening is applied to remove measurements
134 with a cloud fraction of more than 30%. Other details can be found in Theys et al. (2015).

135 Given that missing values are often presented in satellite-retrieved product due to the
136 limitations of retrieval algorithms under adverse environments, it is necessary to
137 evaluate the availability of monthly SO₂ data relative to the entire period. As mapped
138 in Figure S2, there appears to be a substantial fraction of data gaps in western and
139 northeastern China, especially in the winter half year. This can be attributed to snow
140 cover surfaces and high solar zenith angles, which invalidate the measurability. As a
141 result, it may be problematic when sampling western and northeastern China. In
142 contrast, the completeness across eastern parts of China is generally more than 80%
143 regardless of the season, sufficient for inferring the spatial and temporal structures. In
144 what follows, the analysis is mainly confined to the eastern China to avoid issues related
145 to missing data.

146 **2.2 Emission Inventory**

147 The SO₂ emissions at national and provincial level are collected from the China
148 Statistical Yearbook on Environment, which is compiled jointly by the National Bureau
149 of Statistics and Ministry of Environmental Protection. It is an annual statistics
150 publication, with industrial and household emissions listed separately. Currently, this
151 publicly available dataset spans the period from 2003 to 2015, covering 31 provinces
152 in China other than Taiwan, Hong Kong and Macau. Industrial emissions refer to the
153 volume of SO₂ emission from fuel burning and production processes in the premises of
154 enterprises for a given period, while household emissions are calculated on the basis of
155 consumption of coal by households and the sulphur content of coal. Notice that power

156 generation is incorporated into industrial sources and emissions from transportation
157 sources are not reported. This emission inventory released in the official yearbook
158 (OYB for short) has been cited or used in several works, i.e. Li et al. (2017), Yan et al.
159 (2017), Hou et al. (2018), etc.

160 Since a credible emission inventory is the key foundation of this study, the Multi-
161 resolution Emission Inventory for China (MEIC) developed by Tsinghua University (Li
162 et al., 2017; Zheng et al., 2018) has been adopted to verify the OYB inventory as well
163 as to corroborate our findings. The MEIC is a bottom-up emission inventory model
164 including more than 700 anthropogenic sources and then aggregated into five sectors:
165 power, industry, residential, transportation and agriculture. Unlike the OYB estimate,
166 emissions from power plants in MEIC are considered to be a single sector and presented
167 separately. Here, we use province-level emissions from 2003 to 2015, together with the
168 monthly gridded emissions at $0.25^{\circ} \times 0.25^{\circ}$ horizon resolution for the years 2008, 2010,
169 2012, 2014 and 2016. To be in line with the OYB inventory, transportation and
170 agriculture sectors are excluded when calculate summed emission, and the power sector
171 is folded into industrial sector.

172 Figure 1 compares the OYB and MEIC emission inventories in terms of both national
173 and regional scales. In addition, the other two candidates on national annual totals
174 including REASv2 (Kurokawa et al., 2013) and Zhao (Xia et al., 2016) are overlaid.
175 Figure 1a shows that considerable differences exist with regards to the magnitude
176 among the four datasets and in particular OYB emissions are generally lower than those

177 deduced from other inventories. However, their temporal variations are characterized
178 in a very similar manner. As further illustrated in the scatter plot of OYB against the
179 other three (Figure 1b), highly linear clustered markers with correlations above 0.92
180 confirm such temporal consistency. On even smaller regional scale, as shown in Figure
181 1c, high degrees of correspondence between OYB and MEIC overwhelm the whole
182 eastern China, with most correlations exceeding the 0.05 significant level. In
183 comparison, the western China features relatively less agreement, but it is not a major
184 concern in this study.

185 In short, all the datasets capture coherent temporal behaviors, despite the spread in
186 their magnitudes. We emphasize that this study is centered on the fluctuation patterns
187 rather than the magnitude itself. Therefore, the above evidences justify the use of the
188 OYB dataset in the following text. Meanwhile, in order to test whether results were
189 robust to using a different data set, all analyses have been repeated using the MEIC
190 inventory.

191 **2.3 Meteorological Fields**

192 The large-scale meteorological conditions are taken from Japanese 55-year
193 Reanalysis (JRA-55) data, prepared by the Japan Meteorological Agency (Kobayashi
194 et al., 2015; Harada et al., 2016). The variables analyzed include total column
195 precipitable water, horizontal wind and temperature at pressure levels.

196

197 3 General patterns of SO₂ over China

198 3.1 Mean distribution

199 Based on 12 years of SO₂ column data over China, Figure 2a shows the spatial pattern
200 of long-term mean. Overall, SO₂ distribution is of great inhomogeneity in China, with
201 two maximum centers: one is the North China Plain (NCP for short), and the other is
202 Cheng-Yu (CY) district in Southwest China. In particular, SO₂ amount in NCP exceeds
203 1.2 DU. There are two essential causes responsible for high SO₂ loading in the two
204 areas. On the one hand, combined effect of rapid economic and industrial development
205 as well as population growth leads to a high degree of anthropogenic SO₂ emission.
206 Figure 2c and Figure 2d show the emission strengths, defined as emitted SO₂ per unit
207 area, in each province based on OYB and MEIC respectively. Note that in the rest of
208 this paper, the terms “emission” or “emission amount” always refers to “per unit area
209 emission”. It is obvious that the two regions release above 8.0 tons/km² SO₂ per year,
210 which is three times greater than the average level of China. Although OYB exhibits
211 smaller magnitude of emissions than MEIC, the spatial patterns in terms of relative
212 difference across space are generally consistent. On the other hand, as shown in Figure
213 2b, either of the two regions is surrounded or partly surrounded by mountains, which
214 makes it difficult for the pollutants to dissipate.

215 In contrast, over the sparsely populated western part of China, low SO₂
216 concentrations of less than 0.2 DU are observed, except over some provincial capitals.
217 Since western part of China is less affected by human activities, anthropogenic sources

218 of SO₂ are much smaller than natural emissions including emissions from terrestrial
219 ecosystems and oxidation of H₂S to SO₂ (Wang et al., 1999). Between latitude 30-40°N,
220 for example, the SO₂ amount over the eastern regions (110-120°E) are 6-12 times
221 greater than western regions (80-110°E).

222 Besides the NCP and CY regions with highest SO₂ loadings, this study is also
223 interested in Yangtze River Delta (YRD) and Pearl River Delta (PRD), the other two
224 economic mega-urban zones in China. These four identified hotspots NCP, CY, YRD
225 and PRD are outlined in Figure 2a and will be specially examined in the following
226 discussion.

227 **3.2 Seasonal Cycle**

228 The annual total is decomposed into seasonal cycle, as shown in Figure 3. In eastern
229 China, about 35% of the annual totals is taken up by winter, while SO₂ in summer only
230 accounts for 15%; the remaining 50 percent is almost equally divided in spring and
231 autumn. Seasonal variations measured in the fractional contribution are similar within
232 eastern China.

233 To unveil the underlying mechanism, Figure 4 illustrates the annual cycle of SO₂
234 VCDs in relation to sulphur emission, precipitable water and temperature at the four
235 hotspots. Intensive heating during winter in North China raises sulphur release.
236 However, emissions alone are not sufficient to explain the pronounced seasonality of
237 SO₂. The remaining variation is associated with the seasonal change of the
238 meteorological conditions. Temperature and humidity are cold and dry in winter due to

239 the influence of winter monsoon, which jointly weaken the rate of oxidation and wet
240 deposition. Thus, one expects that SO₂ molecules will have a longer lifetime and
241 therefore will accumulate easier. The opposite is true for summer, when chemical
242 reaction is active and wet removal is effective. In summary, both emission and
243 meteorological change explain the seasonality of the atmospheric SO₂ loadings.

244 Due to the climate transition from southern China to northern China, the annual range
245 of SO₂ rises progressively from south to north. NCP has the greatest amplitude of up to
246 1 DU, while there is virtually no annual cycle in PRD. Larger amplitude for SO₂ cycles
247 in NCP arises from the significantly reversed source-sink imbalance between summer
248 and winter. In contrast, the climate in PRD is characterized by smoother transition over
249 the whole year and there is no heating season, which explains the insignificant seasonal
250 variation of SO₂ in PRD. The other two regions CY and YRD have approximately the
251 same amplitude of 0.6 DU, because they are on the same line of latitude.

252 **3.3 Long-term trends**

253 Figure 5 depicts the spatial pattern of linear trends in annual and seasonal SO₂ from
254 2005 to 2016. Overall, apparent downward trends overwhelm most parts of eastern
255 China, while western China has experienced little change. In particular, the most
256 significant reduction occurred in the highly SO₂-polluted regions, with the decreasing
257 rates amounting to 0.1 DU/a. This result suggests that the governments and
258 communities in these economically developed regions have done its best to effectively
259 control environmental pollution, including energy saving, emission cut and adjustment

260 of energy consumption structure, shutdown of the most polluting factories, upgradation
261 of coal quality, etc. Besides, enforcement of environmental protection laws is becoming
262 more and more rigorous (van der A et al., 2017). Therefore, under collaborative efforts,
263 the SO₂ levels in these highly developed regions with high background concentration
264 have been decreasing markedly in the recent decade. Moreover, the pattern correlation
265 between mean (Figure 2a) and trends (Figure 3 top) of SO₂ reaches to -0.77 , implying
266 that the downward rate over China can be summarized into a general rule—the higher
267 the SO₂ loading, the more significant the decrease.

268 Figure 3a-d portrays the long-term trends of SO₂ on seasonal basis. On the one hand,
269 every season has witnessed SO₂ reduction, with the strongest decrease occurring in
270 winter and autumn. Consequently, it can be concluded that the SO₂ decrease in winter
271 and autumn contribute most to the reduction of annual SO₂. On the other hand, the
272 highly SO₂-polluted regions have experienced the most pronounced decrease across all
273 seasons, which is consistent with the annual outcomes. **It is noteworthy to point out that**
274 **a belt of large positive values extend along 40°N in winter (Figure 5a). This feature is**
275 **a known artefact related to the large solar zenith angles at high northern latitudes.**

276 Last, we discuss the trends of the four hotspots interested. Figure 6 depicts the SO₂
277 columns from 2005 to 2016 as a function of year (y-axis) and calendar month (x-axis).
278 The horizontal axis is the month of the year, the vertical axis is the year, and the color
279 is the SO₂ VCD for that month and year. SO₂ VCDs exhibit a decreasing tendency
280 during the last decade, regardless of the time of the year. Quantitatively, SO₂ in NCP,

281 CY, YRD and PRD had undergone an overall downward trend with a rate of 0.062,
282 0.059, 0.046 and 0.055 DU per year, respectively.

283

284 **4 Specific regimes of SO₂ variability and causes**

285 **4.1 Specific regimes of SO₂ variability**

286 The above investigation presents SO₂ patterns and trends across China, but some
287 elusive non-monotonic behaviors are not fully understood. In this section, we aim to
288 detect the specific regimes of SO₂ variability and associated responsible mechanisms.

289 Spatiotemporal regimes of SO₂ over China are mapped by using empirical
290 orthogonal function (EOF) decomposition (Hannachi, 2004), which is a useful tool to
291 reduce the data dimensionality to two dimensions. One dimension represents the spatial
292 structure and the other the temporal dimension. Figure 7 illustrates the leading mode
293 (top) and the corresponding principal component (PC, bottom) obtained from EOF,
294 since only the first mode is statistically well separated. Compared to the first EOF mode
295 explaining 36.8% of the total variance, each of the other modes is characterized by less
296 than 6% contribution and thus discarded. On the one side, the variation of SO₂ is
297 dominated by a spatially uniform feature with large loadings in NCP and CY, suggesting
298 that SO₂ changes would be in the same phase but varying amplitude across the entire
299 region. On the other side, the corresponding PC exhibit overall declines during the 2005
300 to 2016. However, the result does not implicate a simple continuous decrease. In fact,
301 there appears to be a transient increase until a peak and thereafter two sharp drops occur

302 in the periods 2007-2008 and 2014-2016, amid which SO₂ concentrations are under the
303 process of slightly rebounding. In short, the SO₂ variability is characterized by four
304 distinct temporal regimes.

305 Moreover, Figure 8 demonstrates the time series for each province in eastern China,
306 with the segment over 2009-2013 highlighted by red color. It reflects extensive common
307 variation that goes through four stages—that is, a short-lived increasing period at the
308 beginning, a steep drop period during 2007 to 2008, a rebound period of 2009 to 2013
309 and another drastic drop period during 2014 to 2016. Most importantly, it confirms that
310 the SO₂ does not evolve in a monotonic way but shows a striking rebound during 2009
311 to 2013. This pattern is true throughout most of the region, with only two exceptions of
312 Guizhou and Guangdong provinces that had experienced a consecutive decrease since
313 2005.

314 4.2 Causes

315 In this section, we diagnose the likely mechanisms behind the observed SO₂
316 variability. Generally, emissions and meteorological conditions are two main factors
317 that essentially exert influence on atmospheric pollutant load. The impact of changes in
318 emitted SO₂, as the main driving force, is first examined. To this end, the temporal
319 classifications of SO₂ emission for each province based on OYB and MEIC are
320 respectively depicted in Figure 9a and 9b, in which red upward pointing triangle implies
321 non-monotonic decrease with a rebound in the middle whereas persistent decrease is
322 denoted by green downward pointing triangle. In North China except Henan province,

323 both OYB and MEIC datasets show that the emission passed its secondary peak during
324 2009 to 2013. In South China, however, discrepancies between OYB and MEIC emerge
325 in some provinces, namely Jiangxi, Hunan, Guangxi and Guizhou. Even so, we are still
326 confident enough that the majority of South China has witnessed a successive drop in
327 emitted SO₂. In addition, an auxiliary map is presented in Figure 9c showing the slope
328 of the linear regression of MEIC gridded emission over years 2008, 2010 and 2012. We
329 can see that most of North China is subject to a positive rate of change while the
330 opposite holds true over most of South China, which confirms the above findings.
331 Eventually, it comes to conclusion that despite spatially uniformity in temporal-pattern
332 classification of SO₂ VCD (Figure 8), temporal structure of emission demonstrates
333 strong south-north contrast (Figure 9). Therefore, it is advantageous to treat North
334 China and South China separately, as delineated by the dotted line in Figure 9. **Regional**
335 **averaged quantities are estimated as a weighted average by assigning the district area**
336 **as a weight.** In addition, to evade possible contaminations, we have ruled out Henan
337 and Jiangxi provinces in OYB and Henan, Hunan, Guizhou and Guangxi provinces in
338 MEIC.

339 Although we divide the eastern China into north and south blocks, the inter-regional
340 transport cannot be neglected. Therefore, we construct an Effective Emission Index
341 (*EEI*) to account for impacts from both local and remote sources. Here, we directly
342 adopt the results obtained by Zhang et al. (2015), who divided eastern China into three
343 parts North China, Southeast China and Southwest China, and quantified the percent

344 contributions of within-region versus inter-regional transport on sulfate concentrations.
 345 The geographical partition in their work broadly coincides with ours, with the only
 346 difference that South China is further split in two parts. Given that the ratio of Southeast
 347 China to Southwest China is 1.4, we merge the percent contributions over these two
 348 portions via simple conversions. This produces: for North China, within-region SO₂
 349 emission contribute 68% followed by 19% from South China and 13% from other
 350 regions; for South China, within-region emissions provide 66%, while transport from
 351 North China and other regions amounts to 17% and 17% respectively. With these
 352 statistics, the *EEI* is formulated as follows:

$$\begin{array}{ll}
 \text{North China} & \begin{aligned}
 & EEI_1 = 0.68 + 0.19 + 0.13 = 1 \\
 & EEI_m = 0.68 \cdot \frac{N_m}{N_1} + 0.19 \cdot \frac{S_m}{S_1} + 0.13
 \end{aligned} \\
 \text{South China} & \begin{aligned}
 & EEI_1 = 0.17 + 0.66 + 0.17 = 1 \\
 & EEI_m = 0.17 \cdot \frac{N_m}{N_1} + 0.66 \cdot \frac{S_m}{S_1} + 0.17
 \end{aligned}
 \end{array}$$

355 where N and S denote the emission amount in North China and South China respectively,
 356 and subscripts 1 and m the 1st and m^{th} time node respectively. The fundamental
 357 assumptions to derive the formula are that *EEI* is linearly dependent on N and S and the
 358 external contributions remain fixed (without interannual variation). For comparison
 359 purpose, we also define an Emission Index (*EI*) that involves single effect from within-
 360 in region emission, as written below,

$$\begin{array}{ll}
 \text{North China} & \begin{aligned}
 & EI_1 = 1 \\
 & EI_m = \frac{N_m}{N_1}
 \end{aligned} \\
 \text{South China} & \begin{aligned}
 & EI_1 = 1 \\
 & EI_m = \frac{S_m}{S_1}
 \end{aligned}
 \end{array}$$

363 where the notions of symbols are identical to those in *EEI* definition. In the case of
364 large scale, integrating the role of inter-regional transport does not alter the overall
365 pattern, as proved in the following analyses.

366 Figure 10 presents time series and scatter plots of SO₂ VCD and emission with its
367 variants *EI* and *EEI*, and Figure 11 is designed to show the total emission generated by
368 industries and households. These two figures are created based on OYB inventory,
369 while their counterparts obtained from MEIC inventory are shown in Figure S3 and S4
370 in the supplement material. As shown in Figure 10a, the North China features a good
371 correspondence between amount and either *EI* or *EEI*, with linear correlation of 0.9.
372 Time series of emission also indicate the existence of four distinct regimes that are
373 likely to drive the regime of SO₂ VCD directly. This is confirmed by the scatter plot
374 (Figure 10b), in which the points are tightly clustered around the regression line. Based
375 on variance analysis, emission accounts for 81% fraction of SO₂ VCD variation over
376 North China. In parallel, the same procedure relying on MEIC inventory yields nearly
377 identical results, as shown in Figure S3a and S3b. Furthermore, how large do industrial
378 and household sectors respectively contribute to the total trends? Figure 11a and Figure
379 S4a indicate that the industrial emissions play a crucial role in SO₂ VCD variation
380 throughout the whole period, while the influence induced by residential activity is
381 secondary. A more in-depth comparison between OYB and MEIC shows some
382 dissimilarity in household emission: OYB-based household emission acts to offset
383 industrial effect, while opposite function is identified for the MEIC-based one. However,

384 this does not seriously affect the major conclusion, due to the marginal impacts caused
385 by households.

386 The close linear relationship observed in North China is not found in South China,
387 since the two curves appear to become no adherent in Figure 10c and the points in the
388 scatter plot Figure 10d are widely spread around the regression line. Variance analysis
389 suggests that only 45% of SO₂ VCD variability is forced by emissions, suggesting that
390 the SO₂ variations in South China cannot be explained by emission changes alone. This
391 is mainly ascribed to the decoupled pathways of emission and SO₂ VCD during 2009
392 to 2013, as the emission continues to decline but SO₂ VCD witnesses a rebound. MEIC
393 emissions also exhibit a general decreasing tendency in spite of a transient pause
394 embedded, as shown in Figure S3c. Moreover, Figure 11b and Figure S4b suggests that
395 the cuts of industrial and household emissions collectively promote the continuous
396 decrease of total emission in South China, which are different from that in North China.
397 However, the emission decrease in the household sector is differently reported in the
398 OYB and MEIC inventories, the former one showing a sudden shift while the latter
399 displays a gradual decrease. Anyway, it is assured that household emissions in South
400 China have undergone a reduction, irrespective of the exact manner.

401 Why decreasing emissions do not cause a reduction of SO₂ VCD in South China
402 during 2009 to 2013? To answer this question, the atmospheric conditions during 2009
403 to 2013 are compared with those during the rest of the years, as depicted in Figure 12.
404 The period 2009 to 2013 is characterized by prolonged dry conditions in South China

405 with the precipitable water and precipitation being lower than usual (Figure 12a), which
406 weakens wet adsorption and scavenging. At the same time, this period is also associated
407 with relatively weaker wind speed (Figure 12b) and increased static stability (Figure
408 12c, d), reducing the ability of the atmosphere to diffuse leading to the accumulation of
409 SO₂ loads. In brief, unfavorable meteorological conditions produce the observed
410 rebound of SO₂ during 2009 to 2013, despite the continued decrease of emission.

411

412 **5 Conclusions**

413 In this study, the spatiotemporal variability of SO₂ columns over China and the
414 associated driving mechanisms are examined over the past decade. Based on a state-of-
415 the-art SO₂ retrieval dataset recently derived from the OMI instrument, we elaborate on
416 the characteristics of specific SO₂ regimes over China and underlying causes.

417 Climatological SO₂ in China has an uneven spatial distribution in space and time.
418 East China is far more exposed to SO₂ pollution than West China, with two maxima
419 centered in NCP and CY. From analysis of the annual cycles we conclude that 35% of
420 the annual totals are taken up in winter, while SO₂ in summer only accounts for 15%
421 percent. In addition, the annual amplitude of SO₂ rises progressively from south to north.

422 From 2005 through 2016, most of eastern China presents a clear decreasing tendency
423 for SO₂, while western China has experienced little change. Spatially, the decreasing
424 rate is generally enhanced for high SO₂ loads. When computed seasonally, SO₂
425 reductions in winter and autumn contribute most to the reduction of annual SO₂.

426 Four stages of variation are identified by EOF analysis. The first regime (2005-2006)
427 features a transient increasing trend, the second (2007-2008) and the last (2014-2016)
428 regimes show sharp drops, and the third regime (2009-2013) manifests itself by 5-year
429 moderate rebounding. Although temporal regimes of SO₂ are coherent throughout the
430 country, different driving forces are tied to North China and South China. In North
431 China, the atmospheric SO₂ and emission varies essentially in the same way. Therefore,
432 the atmospheric SO₂ variability is primarily associated with the emission variability,
433 which accounts for 81% of the total variance. Further, the emission generated by
434 industrial sector is largely responsible for the atmospheric SO₂ variability. The
435 household emissions appears to **remain uncertain, due to the dissimilarity between OYB**
436 **and MEIC inventories.**

437 SO₂ emissions in South China exhibit a continuous decreasing tendency, due to the
438 **coordinated cuts of industrial and household emissions.** As a result, the role of
439 emissions only contributes one third of the SO₂ variation, primarily owing to the
440 decoupled pathways of emission and atmospheric content during 2009 to 2013 when
441 the emission continues to decline but atmospheric content witnesses a rebound. It is
442 found that such rebound occurs in response to the joint effect of deficient precipitation,
443 weaker wind speed and increased static stability during 2009-2013.

444

445 **6 Future directions**

446 **As enlightened by this study, the spatial and temporal changes of SO₂ regimes over**

447 China in recent decade become clear. However, there is much left to be learned about
448 the responsible driving mechanisms. First, a major obstacle of cause-and-effect relation
449 surveys stems from uncertainties in the current emission inventories. In this study, many
450 facets inferred by OYB and MEIC are convergent, because we look at large spatial scale
451 and long-term general tendency that help filter out or attenuate some uncertainties.
452 However, if the aim is to focus on smaller spatial or temporal scales or on specific
453 sectors, there is still great uncertainty. To overcome these barriers, emission inventories
454 should be further improved and more observational products should be used for
455 comparison. Second, this work investigates the impact of emission, inter-regional
456 transport and meteorology using purely statistical techniques, but finer scale
457 investigations require numerical simulations using coupled chemical-transport models.
458 Third, the analysis presented in Section 4 is constrained to provincial or multi-
459 provincial levels, due to the limitation that only continuous emission data on provinces
460 are gathered at hand. In reality, however, either emission or atmospheric loadings can
461 be quite inhomogeneous within the same region. Therefore, future studies should use
462 both gridded SO₂ VCDs and gridded SO₂ emission inventories.

463

464

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470

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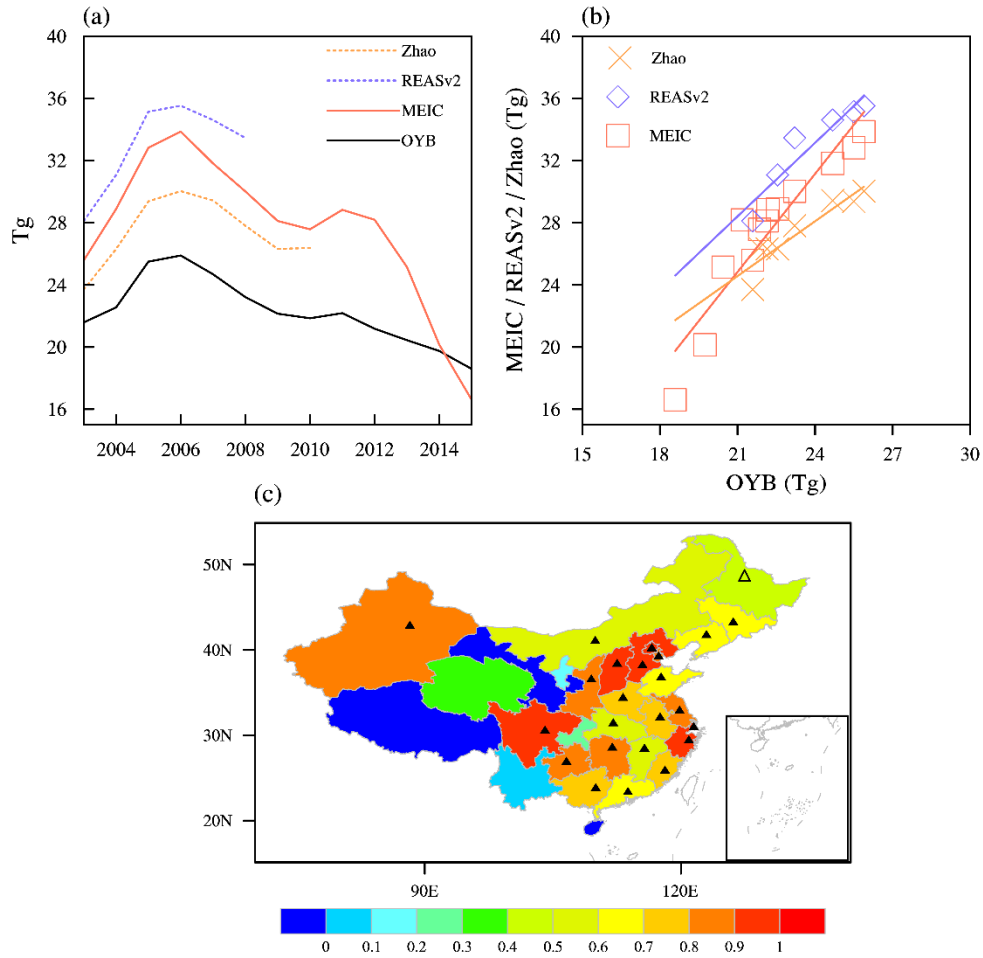
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594 Figure 1 (a) National total SO₂ emissions estimated by OYB (solid black), MEIC (solid red), REAS

595 (dashed blue) and Zhao (dashed orange) between 2003 and 2015. (b) Scatter diagrams and regression

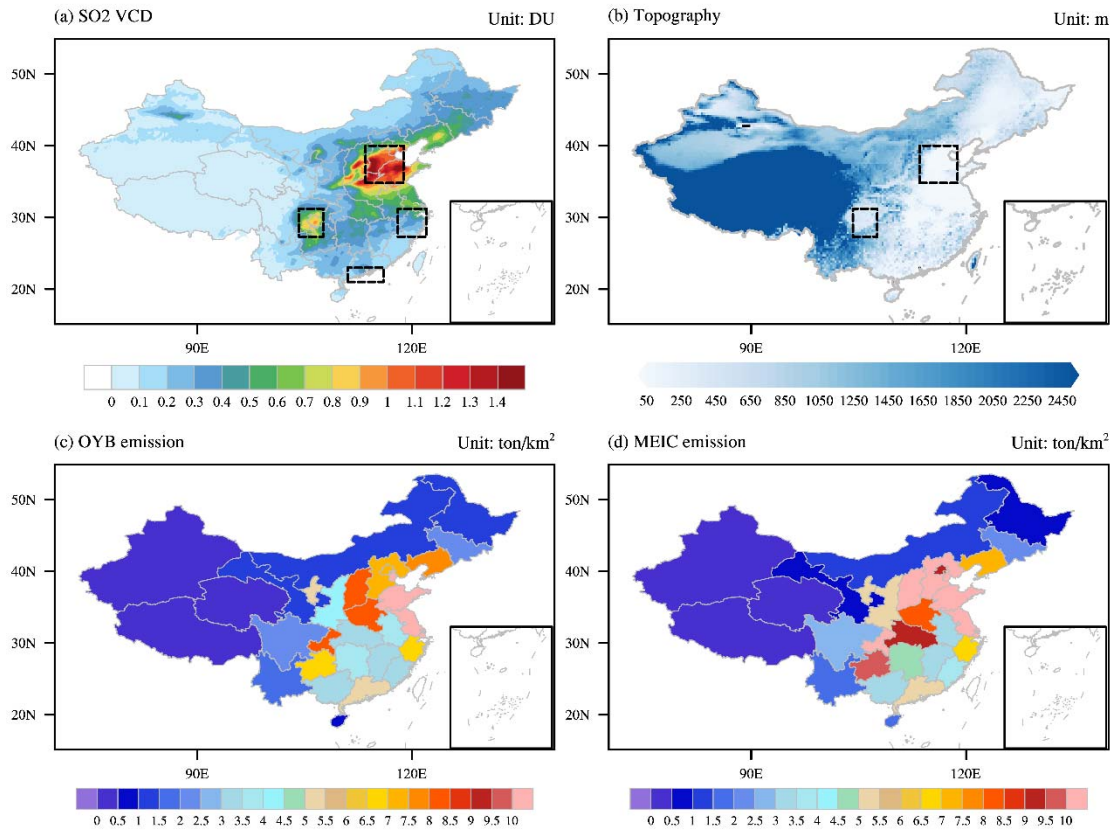
596 lines for OYB estimate (x-Axis) against the other three products (y-Axis). (c) The province-by-province

597 correlations between OYB and MEIC products, with the significance levels of 0.1 and 0.05 are marked

598 by open and filled triangles respectively.

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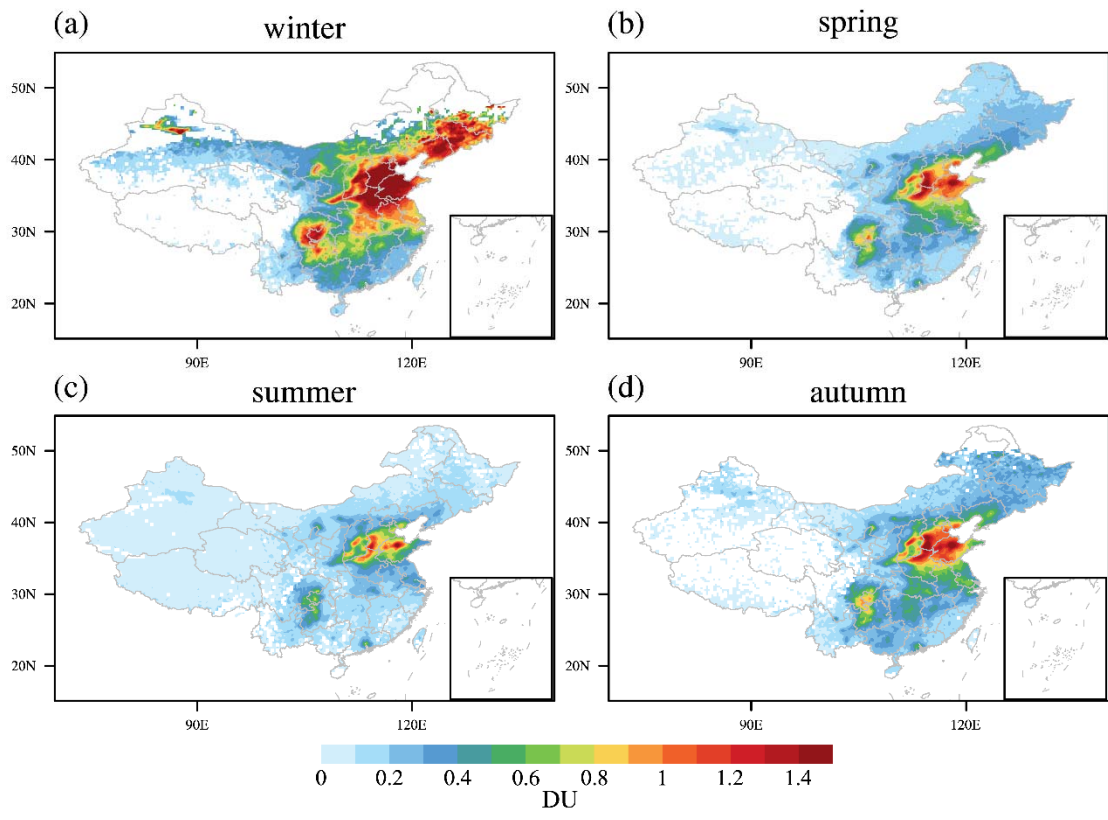
602 Figure 2 (a) Spatial distribution of 12-year (2005–16) averaged tropospheric SO₂ Vertical Column

603 Density (VCD) over China. (b) Topography of China in meters. (c, d) SO₂ emission (ton/km²) among

604 Chinese provinces based on OYB and MEIC.

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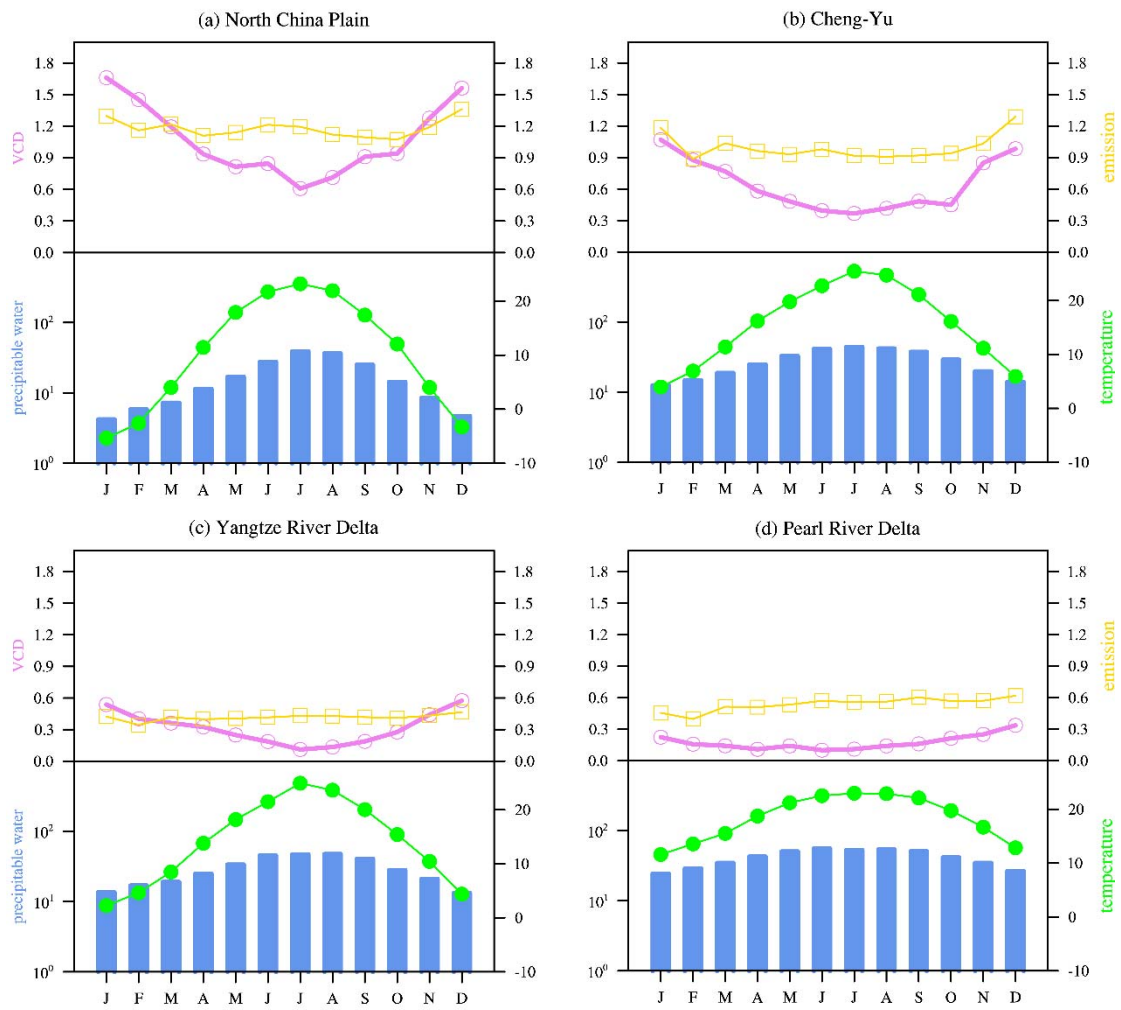
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608 Figure 3 Seasonal tropospheric SO₂ over China: (a) winter, (b) spring, (c) summer and (d) autumn

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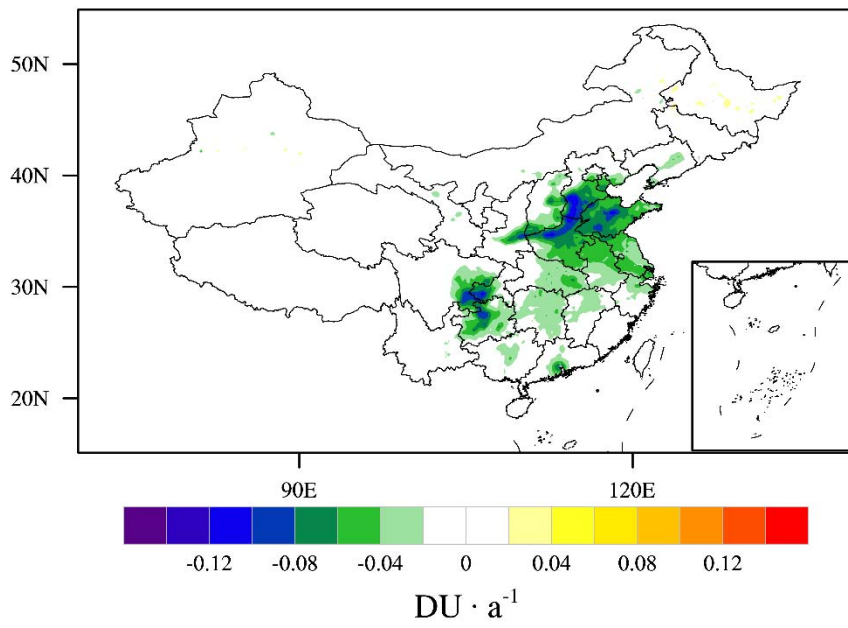
613 Figure 4 Annual cycle of SO₂ VCD (unit: DU, pink line), MEIC SO₂ emission (unit: ton/km², yellow

614 line), precipitable water (unit: kg/m², blue bar) and temperature at 925hPa (unit: °C, green line) for NCP

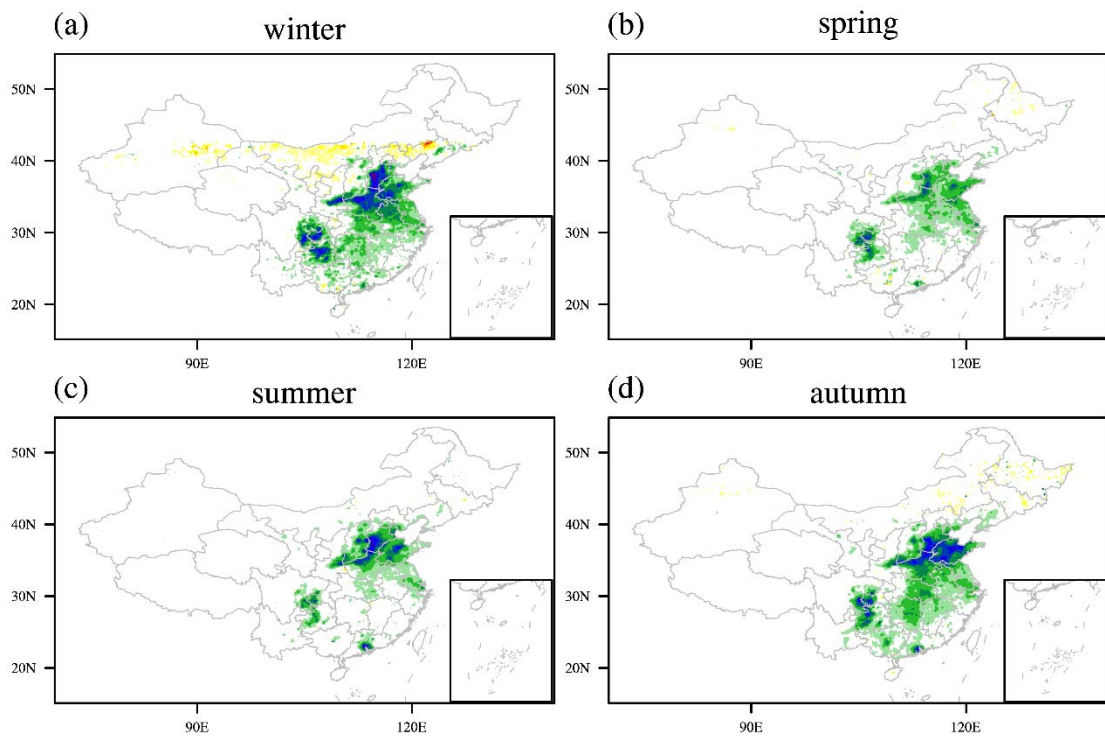
615 (a), CY (b), YRD (c) and PRD (d).

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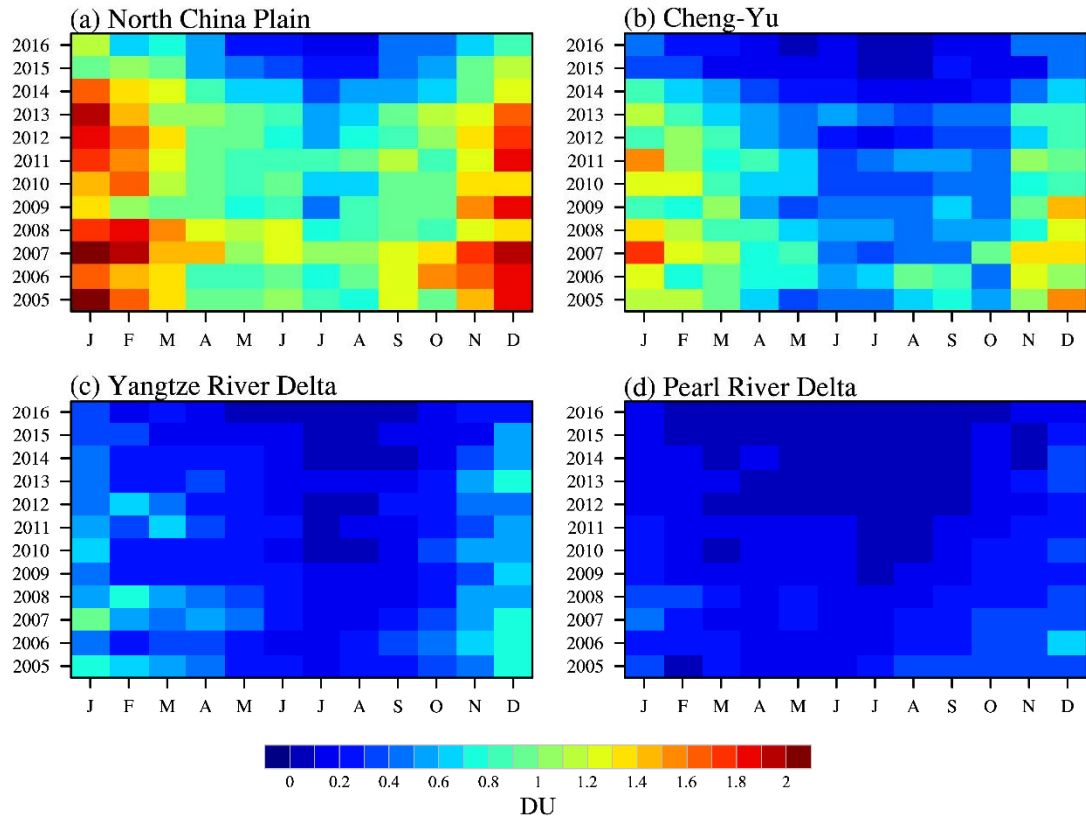
620 Figure 5 Spatial pattern of SO₂ linear trends (2005–16) in annual (Top) and seasonal values (a, b, c,

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d)

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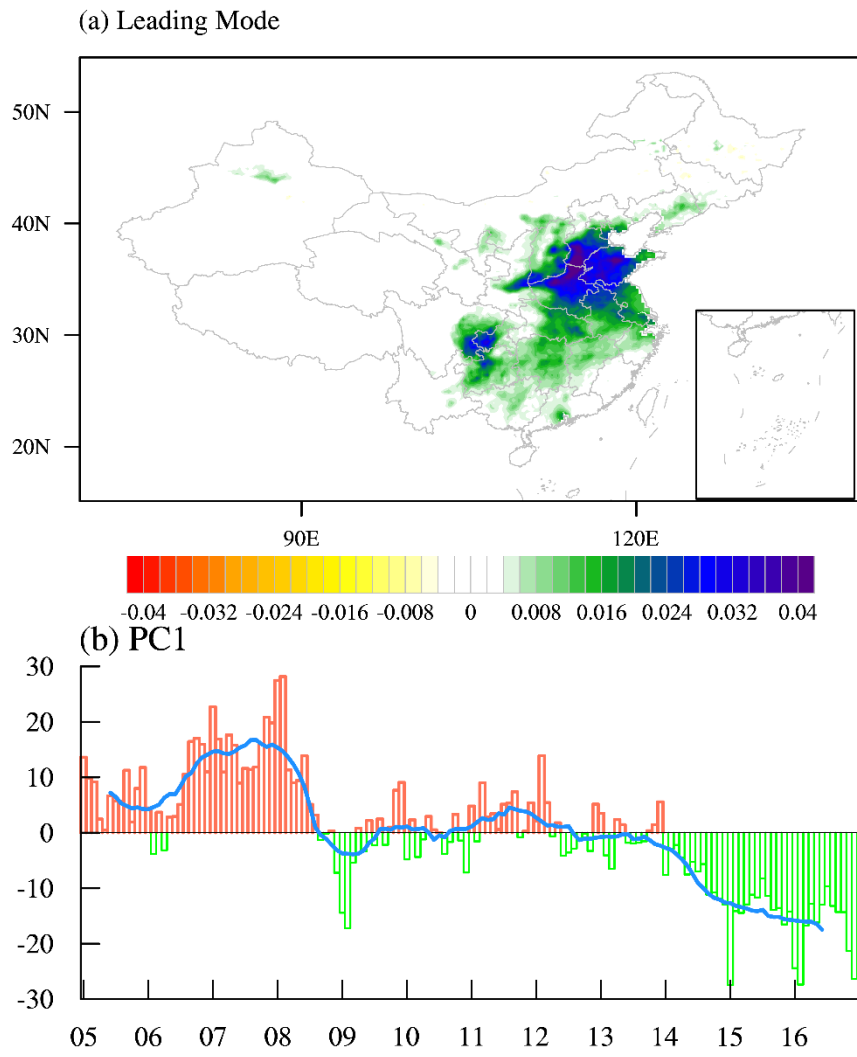
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625 Figure 6 SO₂ amounts from 2005 to 2016 as a function of year (y-axis) and calendar month (x-axis) for

626 NCP (a), CY (b), YRD (c) and PRD (d).

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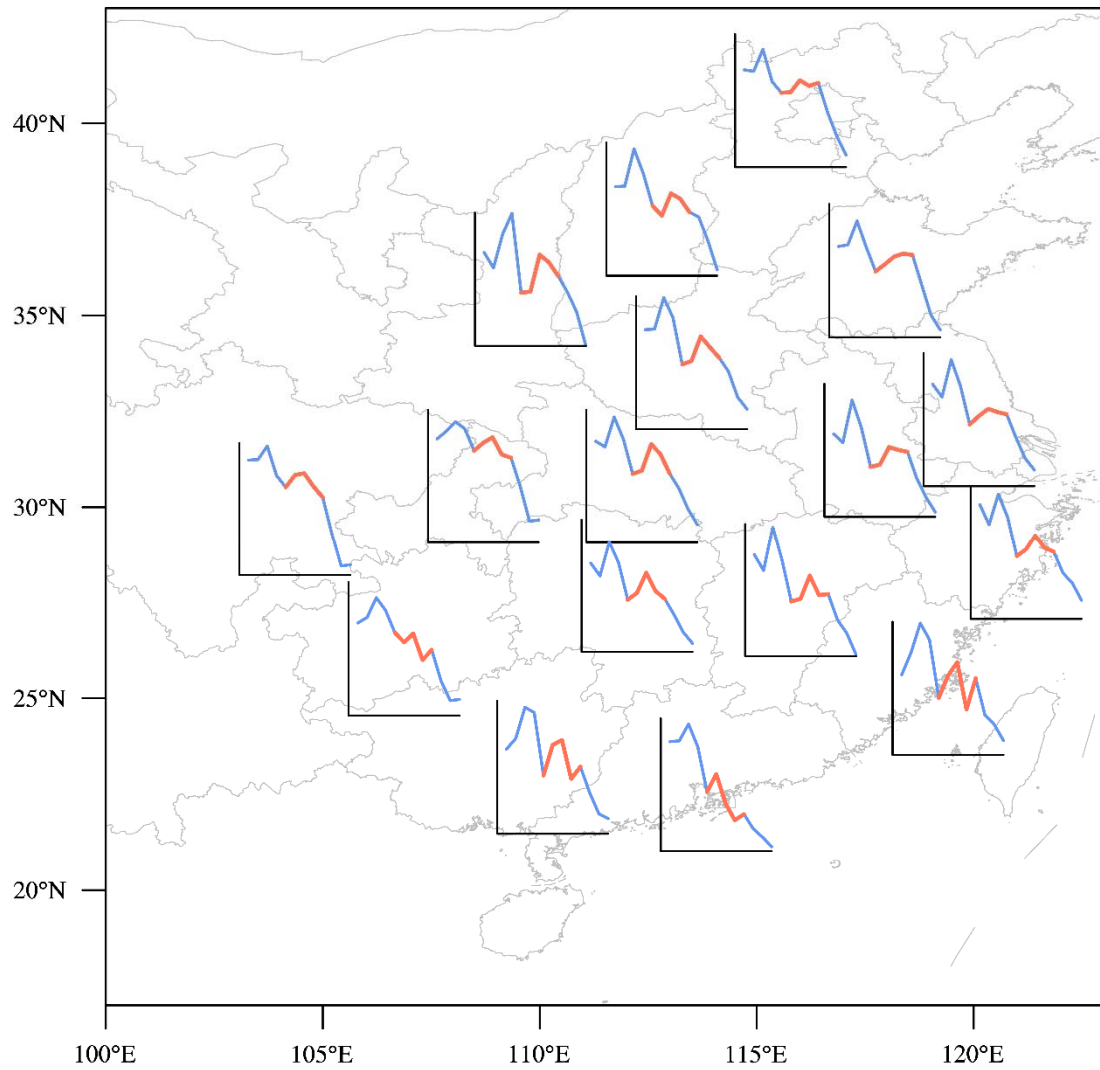
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630 Figure 7 The first leading EOF mode (a) and the corresponding principal components (b)

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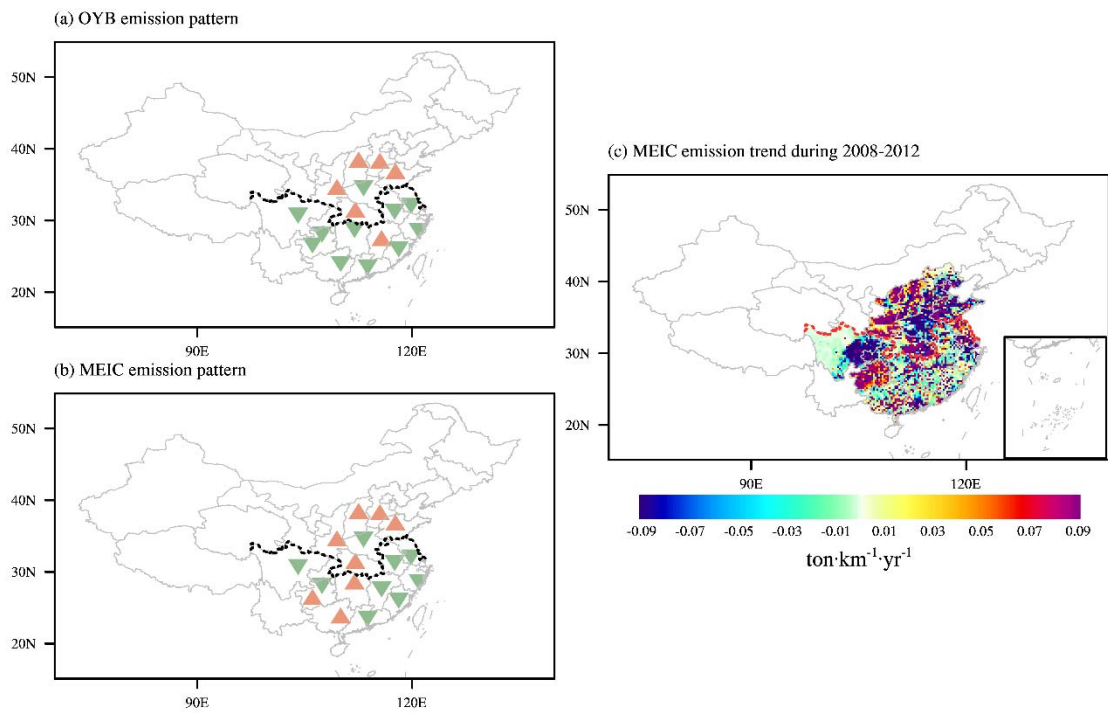
635 Figure 8 Temporal evolution of annual SO₂ (unit: DU) from 2005 to 2016 in each province of eastern

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China, with the segment over 2009-2013 highlighted by red color.

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640 Figure 9 (left panel) Temporal structure classification of SO₂ emission based on OYB and MEIC. Red

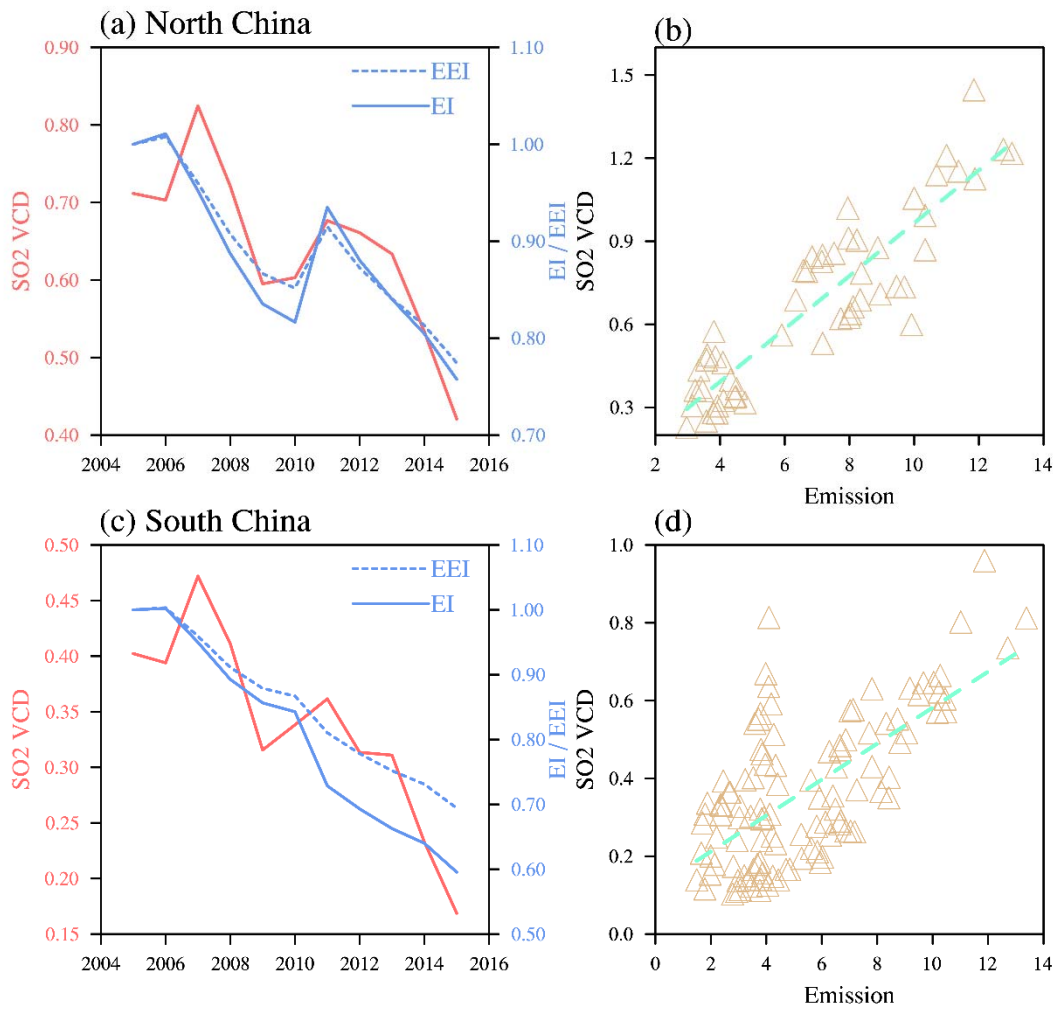
641 upward pointing triangle implies non-monotonic decrease with a rebound in the middle, while monotonic

642 decrease is denoted by green downward pointing triangle. (Right panel) slope of the linear regression of

643 MEIC gridded emission over years 2008, 2010 and 2012. The black or red dotted line delimits the North

644 China and South China.

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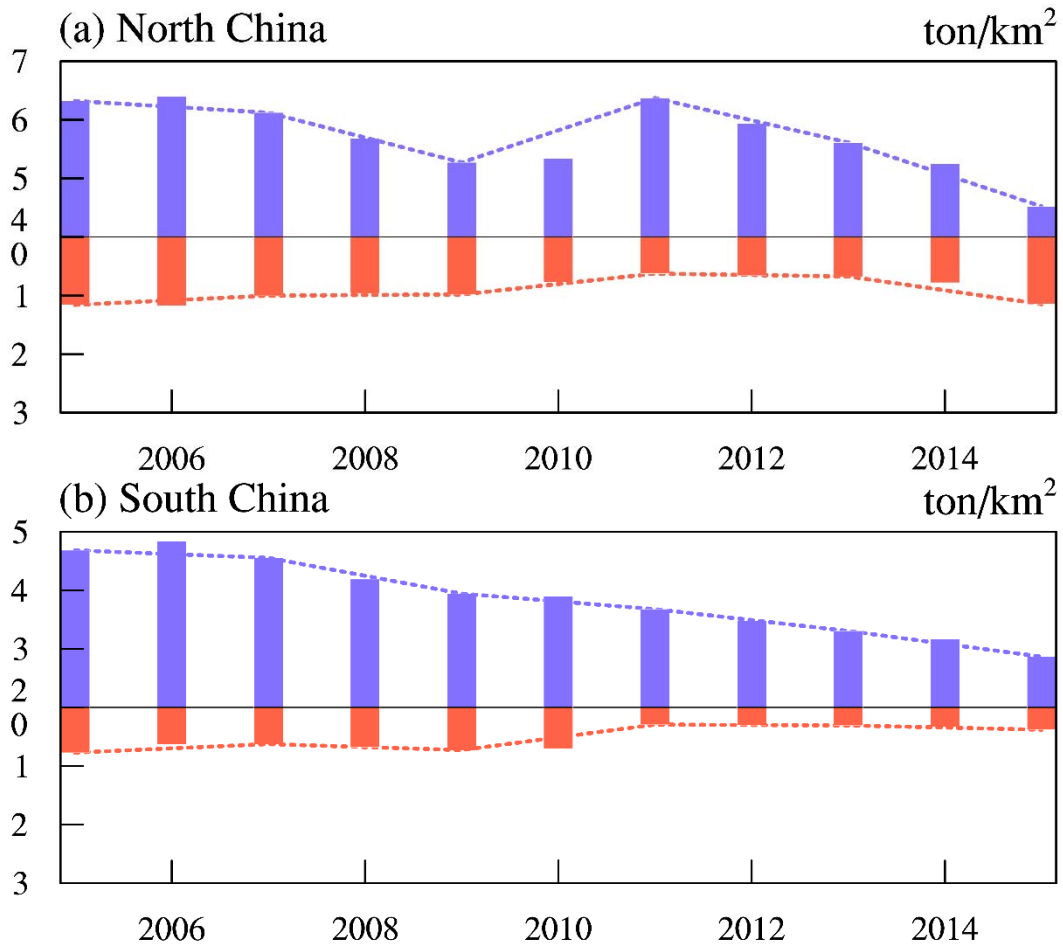
647 Figure 10 Time series plots of SO2 VCD and EI/EEI (a, c), and scatter plots with regression line of

648 SO2 VCD and emission (b, d) for North China (1st Row) and South China (2nd Row). Each marker in b

649 and d corresponds to one year and one province.

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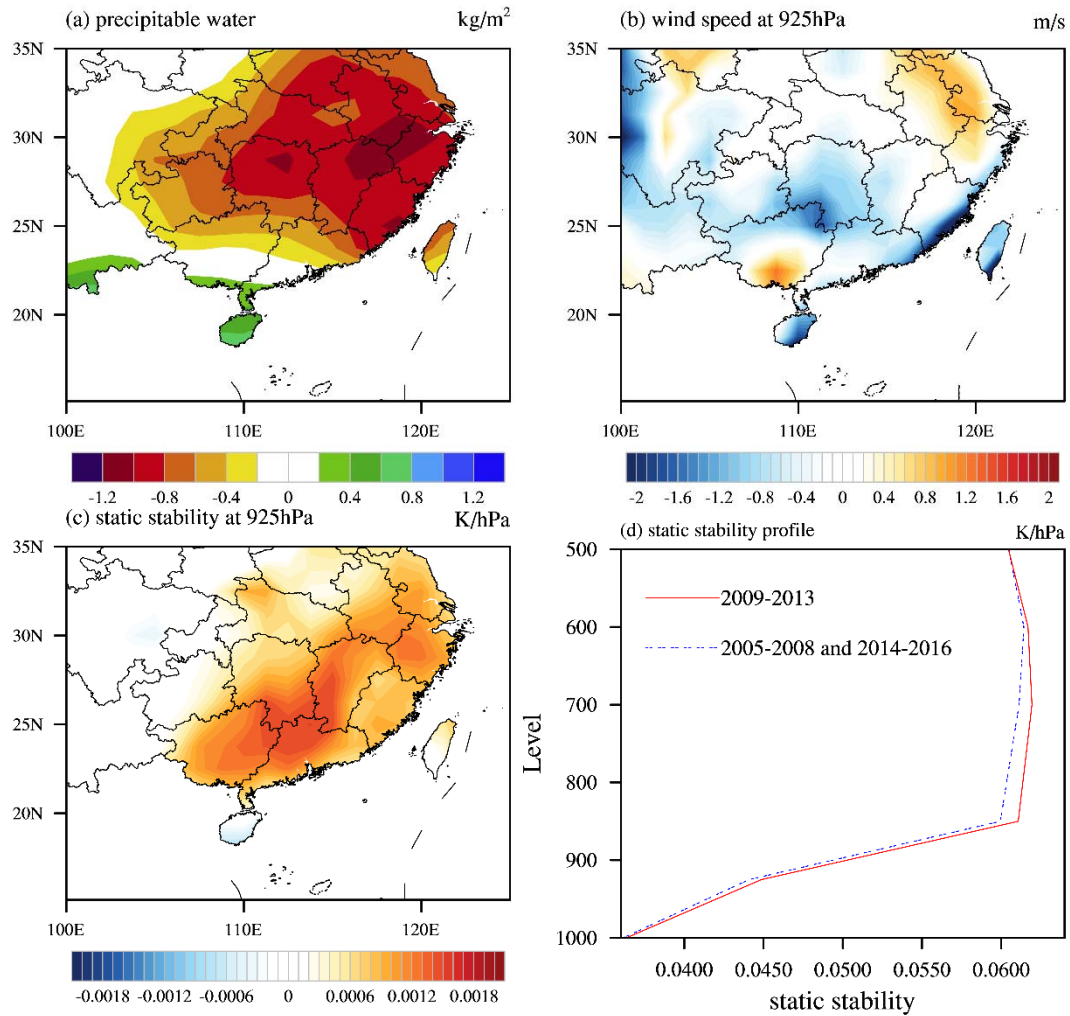
653 Figure 11 Annual SO₂ emission (ton/km²) generated by industries (upward blue bars) and households

654 (downward red bars) in North China (a) and South China (b). Notice that the Y-axis in a positive direction

655 does not start at zero.

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659 Figure 12 Comparison of atmospheric conditions between the period of 2009-2013 and the other years:

660 (a) composite difference in precipitable water (unit: kg/m^2), (b) composite difference in wind velocity at

661 925hPa (unit: m/s), (c) composite difference in static stability at 925hPa (unit: K/hPa), and (d) averaged

662 vertical profile of static stability over the $23\text{-}31\text{N}^\circ$, $105\text{-}122\text{N}^\circ$ rectangle for the two episodes.

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